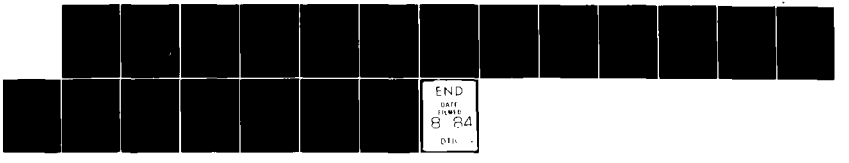
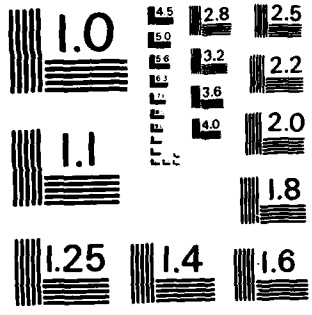
AD-A142 797 ND(3+) BETA'' ALUMINA PLATELET LASER(U) CALIFORNIA UNIV 1/1
LOS ANGELES DEPT OF MATERIALS SCIENCE AND ENGINEERING
M JANSEN ET AL. MAY 84 TR-2 N00014-83-K-0490

UNCLASSIFIED F/G 20/5 NL



END
DATE FILMED
8 84
DTIC



MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS - 1963 - A

(12)

OFFICE OF NAVAL RESEARCH

Contract N00014-83-K-0498

Technical Report No. 2

Nd^{3+} Beta'' Alumina Platelet Laser

by

M. Jansen,* A. Alfrey,* O.M. Stafsudd*
B. Dunn,** D.L. Yang, ** and G.C. Farrington***

Prepared for Publication in
Optics Letters

*Electrical Engineering Department

**Department of Materials Science and Engineering
School of Engineering and Applied Science
University of California
Los Angeles, California 90024

***Materials Science Department
University of Pennsylvania
Philadelphia, Pennsylvania 19104

May, 1984

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER Two (2)	2. GOVT ACCESSION NO. AD-A147 797	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) Nd^{3+} Beta" Alumina Platelet Laser		5. TYPE OF REPORT & PERIOD COVERED Technical Report
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) M. Jansen, A. Alfrey, O.M. Stafsudd, B. Dunn, D.L. Yang, G.C. Farrington		8. CONTRACT OR GRANT NUMBER(s) N00014-83-K-0498
9. PERFORMING ORGANIZATION NAME AND ADDRESS Dept of Materials Science and Engr. University of California, Los Angeles Los Angeles, CA 90024		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
11. CONTROLLING OFFICE NAME AND ADDRESS Office of Naval Research 800 N. Quincy Arlington, VA 22217		12. REPORT DATE May 1984 13. NUMBER OF PAGES 12
14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office) Office of Naval Research Detachment 1030 East Green Street Pasadena, CA 91106		15. SECURITY CLASS. (of this report) Unclassified 15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

This document has been approved for public release and sale; its distribution is limited.

UN

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Pulsed as well as cw laser action has been observed in a new host material for Nd^{3+} ions, Na^{+} beta" alumina. The unusual characteristics of the material and its known physical properties are briefly described. Absorption spectra, fluorescence lifetime, laser-pulse shape, and peak emission wavelength are presented as experimental evidence, and comparisons are made with other Nd^{3+} host materials. Some exciting new possibilities for integrated-optics applications are explored.

DD FORM 1 JAN 73 1473 EDITION OF 1 NOV 65 IS OBSOLETE

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

Nd^{3+} BETA" ALUMINA PLATELET LASER

M. Jansen, A. Alfrey, O.M. Stafsudd

Electrical Engineering Dept. at U.C.L.A.

B. Dunn and D. L. Yang

Materials Science Dept. at U.C.L.A.

G.C. Farrington

Materials Science Dept. at the University of Pennsylvania

ABSTRACT

Pulsed, as well as c.w. laser action has been observed in a new host material for Nd^{3+} ions, Na^{+} beta" alumina. The unusual characteristics of the material and its known physical properties are briefly described. Absorption spectra, fluorescence lifetime, laser pulse shape, and peak emission wavelength are presented as experimental evidence, and comparisons are made to other Nd^{3+} host materials. Some exciting new possibilities for integrated optics applications are explored.

1. INTRODUCTION

Sodium beta" alumina ($Na_{1+x}\ Mg_x\ Al_{11-x}\ O_{17}$) is a non-stoichiometric sodium aluminate known for its ability to transport sodium ions (Na^+). The structure, properties and applications of the material have been well reviewed.[1-3] Its structure consists of "spinel blocks" of close-packed Al^{3+} and O^{2-}, separated by loosely packed planes containing Na^+ and O^{2-}. The open space in the conduction planes allows rapid Na^+ migration in two-dimensions.

Farrington and Dunn have shown that both divalent and trivalent cations may exchange for the Na^+ ions.[4,5] Complete replacement was obtained for various divalent cations such as Ca^{2+}, Sr^{2+}, Ba^{2+}, Pb^{2+} and Cd^{2+}, and trivalent cations such as Gd^{3+}, Nd^{3+}, and Eu^{3+}.

Ion transport phenomena is used to introduce the optically active ions after crystal growth. As a Nd^{3+} host, the solid exhibits some spectral anomalies, which are discussed in more detail in section 5. Laser action has been observed in both pulsed and c.w. modes of operation.

2. MATERIAL SYNTHESIS

The ion exchange procedure was analogous to that described previously .[4] Single crystals of Na^+ beta"

alumina were placed in a molten salt mixture (55% NaCl-45% $NdCl_3$) at 600 °C for various periods of time. The amount of Nd^{3+} exchange was determined gravimetrically and depended upon the length of immersion time. The crystals prepared for this study contained a Nd^{3+} concentration of ca. $7x10^{20}$ cm^{-3} which is substantially greater than the $1x10^{20}$ cm^{-3} generally employed for Nd:YAG. Larger Nd^{3+} levels can be obtained by using pure $NdCl_3$ melts and achieving complete exchange (ca. $1.5x10^{21}$ cm^{-3}),[5] however, this process etches the crystal and lessens its optical quality. The low melting flux chosen for this work avoids these difficulties. Thus, in the experimental results reported below, Nd^{3+} ion exchange was not complete and the conduction plane contained Na^+ (ca. $3x10^{21}$ cm^{-3}) as well as Nd^{3+}. It is interesting to note that x-ray diffraction studies indicate that trivalent and Na^+ ions occupy different sites in the beta" alumina conduction plane (9d and 6c sites, respectively; Wyckoff notation).[3,6]

3. INDEX OF REFRACTION

The index of refraction of highly doped crystals was determined from multiple angle ellipsometry measurements. A Rudolph Research ellipsometer, with a HeNe laser source was

used in obtaining the preliminary data, which were later analyzed by computer techniques[7] to yield a 1.79 ordinary index of refraction for the Nd^{3+} doped Na^{+} beta"-alumina sample. The ellipsometry data also revealed a high degree of anisotropy in the measured crystals, but because of the very small size of the crystals available at this time, the extraordinary index of refraction (corresponding to electric fields perpendicular to the large face of the platelets) could not be determined.

4. LIFETIME MEASUREMENT

The fluorescent liftimes of several partially exchanged crystals, with doping densities varying between $2x10^{19}$ cm^{-3} and $1.5x10^{21}$ cm^{-3}, are shown in figure 1 as a function of the doping concentration. It can be seen that the measured fluorescent lifetimes vary between 350 and 400 $\pm$ 50 microseconds, which is similar in range to the values reported in the literature for other Nd^{3+} hosts. One notable exception is that no fluorescent lifetime reduction is observed at high doping concentrations, as is the case in Nd^{3+}-YAG[8].

5. PRELIMINARY SPECTRAL ANALYSIS

The absorption spectra was measured on a Cary IR-14 specrophotometer using both rapid and very slow scans to allow accurate integration of the data (see fig.2). When compared to other known Nd^{3+} spectra, such as those of Nd^{3+}:YAG or glass, the samples were found to exhibit an anomalously high absorption intensity in the 580 nm region of the spectrum. To illustrate this point, we calculated the oscillator strengths (as defined by Hoogschagen[10] and Judd[11]) for the various transitions in Nd^{3+}-YAG, from Krupke's [9] tabulated values of "integrated absorption coefficients". For the absorption line centered at 580 nm, the oscillator strength for a Nd^{3+}-YAG sample having a Nd^{3+} concentration of 1.06×10^{20} cm^{-3} was found to be 7.05×10^{-6}. For comparison, a 3.1×10^{-2} cm thick Nd^{3+} beta" alumina sample, having a 4×10^{20} cm^{-3} Nd^{3+} concentration, yielded an oscillator strength of 60.6×10^{-6} for the 580 nm band.

The general spectral behavior of the crystal is still under investigation, and it is expected to yield additional information pertaining to the lattice structure of the material, and the location of ion sites. A theoretical model accounting for the anomalous absorption is being developed, and will be published elsewhere.[12]

A further experiment was performed in order to obtain an excitation spectrum, and to measure the relative fluorescent emission in the infrared. A heavily doped (Nd^{3+}= $4.7x10^{20}$ cm^{-3}) beta"-alumina crystal was pumped with a commercial nitrogen pumped dye laser system. The four different dyes used were tuned over most of the visible spectrum, from 453 nm to 643 nm. The excitation spectra and the corresponding fluorescent output centered at around 1.06 microns are shown in table 1. The results indicate the existence of a very efficient excitation band at 583.9 nm, not far from the peak of the anomalous absorption region.

When broken along the natural cleavage planes, the crystals could be separated into very thin sheets. This suggested the possibility of producing waveguide lasers, with the feedback being provided by Fresnel reflections at the cleaved crystal ends. Despite a lack of preliminary preparation, such as polishing, the end faces were found to be surprisingly flat, and of good optical quality. The 8% reflectivity at each end, based on Fresnel reflection alone, resulted in high loss resonator structures.

Laser action was obtained in both pulsed and c.w. modes of operation, in spite of the low feedback level. Sufficiently high inversion levels, and accordingly high

gains were obtained in crystals with large Nd^{3+} doping concentrations, by efficiently pumping them within the narrow excitation band centered at 583.9 nm. In all cases the pump energy was launched in the direction of the platelet laser emission (longitudinal configuration), and the estimated pumping volume varied from $5x10^{-3}$ cc in the pulsed experiments to $5x10^{-4}$ cc in the c.w. experiments. Since the feedback in the cavity was provided by Fresnel reflections alone, the lower limit for the small signal gain at threshold can be calculated to be of the order of 5 per cm.

6. PULSED OPERATION OF THE PLATELET LASER

Crystals ranging from .25 to .5 cm in length and from .05 to .25 mm in thickness were pumped by a National Research Group nitrogen - dye laser system. The nitrogen laser was rated at 0.7 megawatts peak pulse power, at a repetition rate of up to 60 Hz, and a pulse duration of of 5 nanoseconds FWHM. A 10% dye laser optical conversion efficiency was achieved at 590 nm, with Rhodamine 6G dye dissolved in ethyl alcohol. A linearly variable attenuator

was used to lower the pump power, and the output was measured with a S1 photomultiplier (RCA 7102).

The crystal was oriented so that the pumping, as well as the waveguiding action would occur along the natural cleavage planes. The normalized pump power vs. crystal output in the infrared is illustrated in fig.3. A threshold behavior is observable, and in fact, saturation effects become noticeable at high pumping levels. Temporal measurements were obtained and are shown in fig. 4. Pulse widths of the order of 600 nanoseconds (FWHM) were obtained.

Relaxation oscillations are also visible on the pulse trace. Their low amplitude can be attributed to the high cavity loss.

7. CW OPERATION OF THE PLATELET LASER

C.w. laser action has been observed when pumping Nd^{3+} doped beta" alumina crystals with a Spectra-Physics argon ion pumped dye laser system. Up to one watt of 580 nm radiation was focused on the sample, and a spotsize of approximately 0.05 mm was obtained by using a 10X microscope objective.

A S1 photomultiplier (RCA 7102), in conjuction with either a PAR model 124 lock-in amplifier, or an

oscilloscope, were used in the detection of the infrared signal. As in the pulsed case, a very convincing threshold behavior is apparent, as illustrated by figure 3. The smooth threshold transition can be attributed to the high level of spontaneous emission present at the output.

Waveguiding was established in samples varying from 50 to 100 microns in thickness. Due to the large sample thickness, a very large number of modes were allowed to oscillate. Pinhole scans of the far field pattern indicated a very complex mode structure, and confirmed the existence of a large numerical aperture.

Preliminary spectral measurements validated the existence of an infrared emission peak at 1.059 microns, and indicated the presence of strong inhomogeneous broadening.

8. CONCLUSION

Lasing has been achieved in both pulsed and c.w. modes of operation in a new active material, i.e. Nd^{3+} doped beta" alumina. The high cavity loss regime, established by the high transmissivity of the two cleaved ends, had to be offset by a sufficiently high gain. This was achieved by pumping the sample in an efficient manner, and by providing a substantial Nd^{3+} doping concentration.

The ability of preferentially doping various regions of the sample with active ions is unprecedented, and this may be accomplished because of the ion transport behavior of the material. Thus, an obvious application would be in the making of integrated, optically active devices. Many configurations are possible, suggesting a number of very interesting surface wave devices, which, in turn, could be used in areas ranging from communications to optical computing. Waveguiding has already been demonstrated in relatively thick samples, and the diffusion process is being refined, to allow for the construction of more complex surface wave structures. An added advantage lies in the output wavelength of such devices when doped with Nd^{3+}, since it would permit long distance optical fiber transmission with a relatively low loss.

In order to minimize the size of an integrated optics package, pumping by semiconductor lasers, although not as efficient as the present pumping scheme, should be considered. There are several examples in the literature[13-17], where such pumping has resulted in lasing for other Nd^{3+} host materials.

Other ions can also be exchanged with the sodium in the beta" aluminas. The ongoing investigation into the optical

properties of such optically active ions, may very well result in the discovery of new laser materials. The diffusion of different rare earth ions may be also be used as very sensitive optical probes, to identify localized ionic distributions, and to gain further understanding into the way energy is being exchanged in such structures.

The authors wish to acknowledge the contributions of Prof. E.Y. Wong of the U.C.L.A. Physics Dept. for his interpretation of the absorption spectrum. This work was supported in part by the Office of Naval Research.

REFERENCES

1. J.T. Kummer, "Beta-alumina electrolytes", Progr. Solid State Chem.,7, 141 (1972).
2. J.B. Bates, J-C. Wang, and N.J. Dudney, "Solid electrolytes - the beta aluminas", Phys. Today,35, 46-53 (1982).
3. J.P. Boilot, G. Collin, Ph. Colomban, R. Comes, "X-ray-scattering study of the fast ion conductor beta"-alumina",Phys. Rev. B, 22, 5912-5923 (1980).
4. G.C. Farrington and B. Dunn,"Divalent beta"-aluminas: high conductivity solid electrolytes for divalent cations", Solid State Ionics, 7, 267-281 (1982).
5. B. Dunn and G.C. Farrington, "Trivalent ion exchange in beta"-alumina", to be published in Solid State Ionics.
6. W. Carrilo-Cabrera, J.O. Thomas and G.C. Farrington, "The ionic distribution in trivalent Gd^{3+} Beta" Alumina, to be published in Solid State Ionics.
7. M.E. Pedinoff and O.M. Stafsudd, "Multiple angle ellipsometric analysis of surface layers and surface layer contaminants", Applied Optics, 21, 518-521 (1982).

8. A.A. Kaminskii, Laser Crystals, Springer-Verlag Ed., 327 (1981).

9. W. Krupke, IEEE J. Quantum Elec., QE-7, 153-159 (1971)

10. J. Hoogshagen and C.J. Gorter, Physica, 14, 197-206 (1948)

11. B.R. Judd, Phys. Rev., 127, 750-761 (1962)

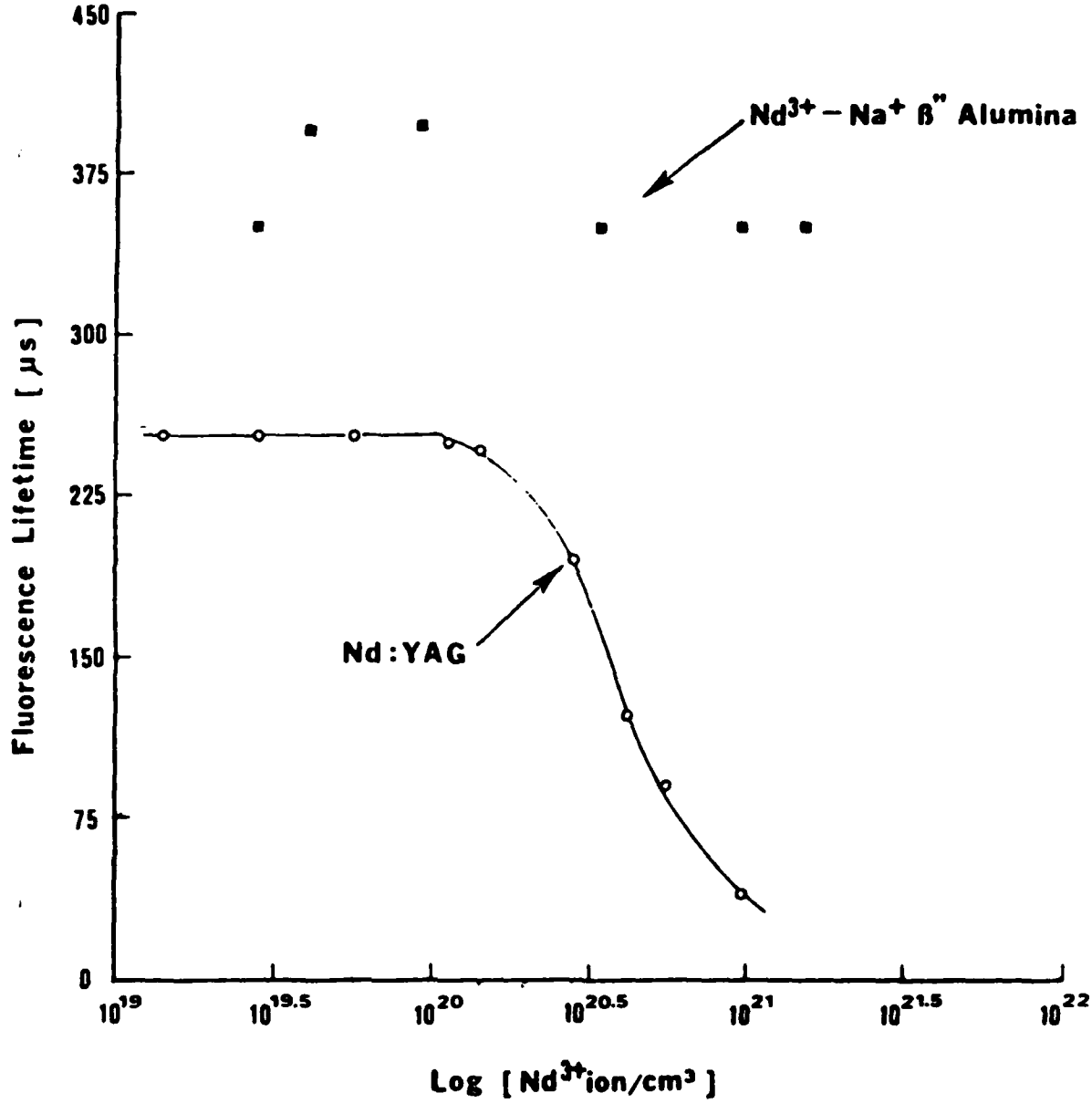
12. E. Wong, private communication.

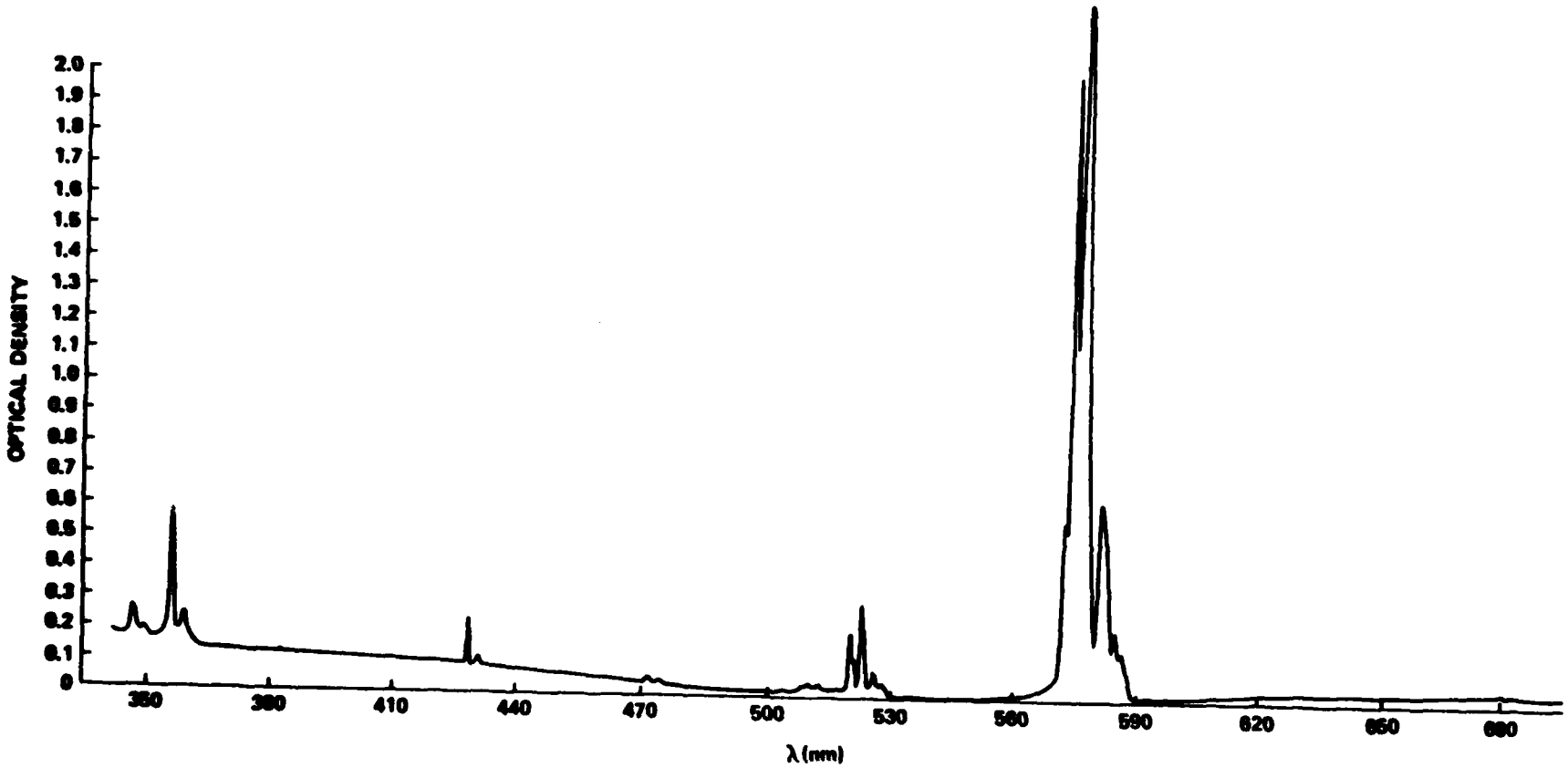
13. R. B. Chesler and S. Singh, "Performance model for end-pumped miniature Nd:YAG lasers", J. Appl. Phys., 44, 5441-5443 (1973).

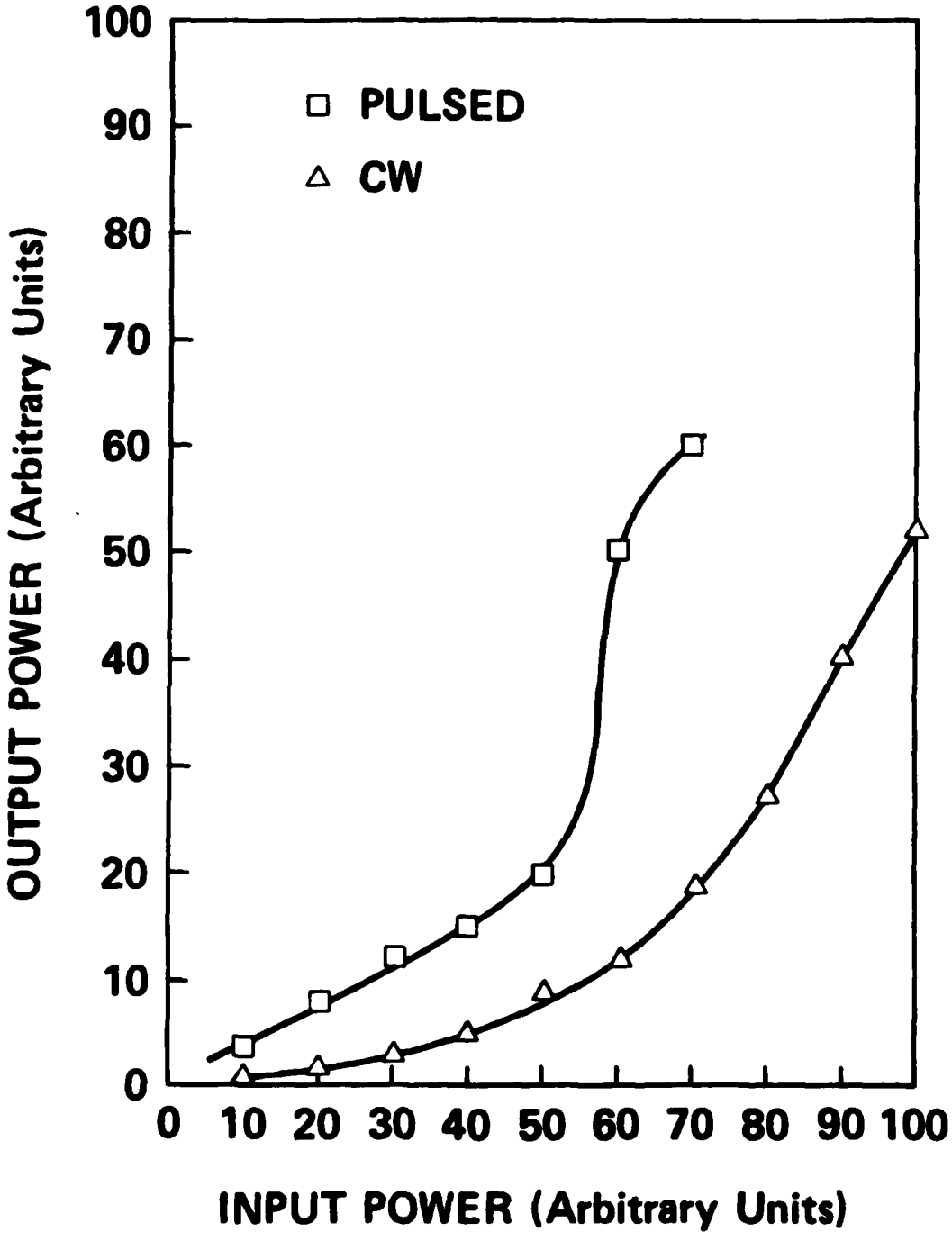
14. M. Saruwatari and T. Kimura, "LED pumped lithium neodymium tetraphosphate lasers", IEEE J. Quantum Electron, QE-12, 584-591 (1976).

15. J. Stone, C.A. Burrus and A.G. Dentai, "Nd:YAG singlr crystal fiber laser: room-temperature cw operation using a single LED as an end pump", Appl. Phys. Lett., 29, 37-39 (1976).

16. J. Stone and C.A. Burrus, "Nd:Y_2O_3 single-crystal fiber laser: room temperature cw operation at 1.07 and 1.35 microns wavelength",J. Appl. Phys., 49, 2281-2287 (1978).


450
375
300
225
150
75
0
Fluorescence Lifetime [μs]
Nd3+ – Na+ ß" Alumina
Nd : YAG
10^19
10^19.5
10^20
10^20.5
10^21
10^21.5
10^22
Log [Nd3+ ion/cm3]


OPTICAL DENSITY
2.0
1.9
1.8
1.7
1.6
1.5
1.4
1.3
1.2
1.1
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
350
380
410
440
470
500
530
560
590
620
650
680
λ (nm)


PULSED
CW
OUTPUT POWER (Arbitrary Units)
INPUT POWER (Arbitrary Units)
0
10
20
30
40
50
60
70
80
90
100

OUTPUT POWER (ARBITRARY UNITS)
0
10
TIME (0.1 μsec/div.)

END

DATE
FILMED

8 84

DTIC